

WE CLAIM:

1. An apparatus for use in the detection of an analyte by electrochemiluminescence comprising an electrode and a counterelectrode, said electrode or counterelectrode having immobilized on a surface thereof a predetermined amount of a reagent capable of binding an electrochemiluminescent labelled species or a binding partner of an electrochemiluminescent labelled species.
2. An apparatus for use in the detection of an analyte by electrochemiluminescence comprising an electrode and a counterelectrode, said electrode or counterelectrode having immobilized on a surface thereof a predetermined amount of a reagent capable of directly or indirectly binding an electrochemiluminescent labelled species.
3. An apparatus for use in the detection of an analyte by electrochemiluminescence comprising an electrode, said electrode having immobilized on a surface thereof a predetermined amount of a reagent capable of binding a component of a binding electrochemiluminescence assay.
4. An apparatus as recited in claim 3 wherein said electrode is the working electrode.
5. An apparatus as recited in claim 3 wherein said electrode is the counterelectrode.
6. An apparatus as recited in claim 3 wherein the immobilized reagent and electrode complex is substantially permissive to electron transfer between said electrode and an electrochemiluminescent label.

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7. An apparatus as recited in claim 3 wherein said electrode is comprised of a porous material.

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8. An apparatus for detection of an analyte by electrochemiluminescence comprising a porous electrode and a counterelectrode.

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9. An apparatus for use in the detection of an analyte by electrochemiluminescence comprising a working electrode and a counterelectrode, said working electrode having immobilized on a surface thereof and in electrochemical contact therewith, a predetermined amount of a reagent capable of binding a component of a binding electrochemiluminescence assay.

10. An apparatus for use in the detection of an analyte by electrochemiluminescence comprising a working electrode and a counterelectrode, said counterelectrode having immobilized on a surface thereof and in electrochemical contact therewith, a predetermined amount of a reagent capable of binding a component of a binding electrochemiluminescence assay.

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11. An apparatus for use in the detection of an analyte by electrochemiluminescence comprising an electrode, a counterelectrode and a porous matrix in electrochemical contact with said electrode, said porous matrix containing on a surface thereof a predetermined amount of a reagent capable of binding a component of a binding electrochemiluminescence assay.

12. An apparatus for use in the detection of an analyte by electrochemiluminescence comprising an electrode and a plurality of discrete binding domains, said binding domains being

capable of binding a component of a binding electrochemiluminescence assay.

13. An apparatus for use in the detection of an analyte by electrochemiluminescence comprising a working electrode and a plurality of discrete binding domains each containing a predetermined amount of a reagent capable of binding a component of a binding electrochemiluminescence assay

14. An apparatus as recited in claim 13 further comprising a counterelectrode.

15. An apparatus as recited in claim 13 further including a counter electrode, sample delivery means and light detection means.

16. An apparatus for use in the detection of an analyte by electrochemiluminescence comprising

(a) an electrode,

(b) a plurality of discrete binding domains each containing a predetermined amount of a reagent capable of binding a component of a binding electrochemiluminescence assay,

(c) a counterelectrode,

(d) means for sample delivery to said binding domains,

(e) means for triggering electrochemiluminescence, and

(f) means for electrochemiluminescence detection

17. An apparatus for use in the detection of an analyte by electrochemiluminescence comprising

(a) an electrode,

SM 25. An apparatus as recited in claim 17 wherein said support is a porous matrix, said matrix being positioned between said electrode and counterelectrode.

SM 26. An apparatus for use in the detection of an analyte by electrochemiluminescence comprising

- (a) an electrode,
- (b) a plurality of discrete binding domains each containing a predetermined amount of a reagent capable of binding a component of a binding electrochemiluminescence assay,
- (c) a counterelectrode,
- (d) means for sample delivery to said binding domains,
- (e) means for triggering electrochemiluminescence, and
- (f) a plurality of means for electrochemiluminescence detection.

SM 27. An apparatus as recited in claim 26 wherein said plurality of means for electrochemiluminescence detection is used to image one or more of said binding domains.

SM 28. An apparatus as recited in claim 27 wherein at least one of said plurality of detection means detects electrochemiluminescence from one binding domain.

SM 29. An apparatus as recited in claim 27 wherein said plurality of means for electrochemiluminescence detection is a CCD array or a diode array.

SM 30. An apparatus for use in the detection of an analyte by electrochemiluminescence comprising an electrode

having immobilized on a surface thereof a plurality of discrete binding domains, said binding domains being capable of binding a component of a binding electrochemiluminescence assay.

31. An apparatus for use in the detection of an analyte by electrochemiluminescence comprising an electrode having immobilized on a surface thereof a plurality of discrete binding domains each containing a predetermined amount of a reagent capable of binding a component of a binding electrochemiluminescence assay.

32. An apparatus for use in the detection of an analyte by electrochemiluminescence comprising an electrode having immobilized on a surface thereof a plurality of discrete binding domains, said binding domains containing an electrochemiluminescent label.

33. An apparatus for use in the detection of an analyte by electrochemiluminescence comprising an electrode having immobilized on a surface thereof a plurality of discrete binding domains, said binding domains being capable of generating a plurality of electrochemiluminescence signals; and a counterelectrode.

34. An apparatus for use in the detection of an analyte by electrochemiluminescence comprising a support having immobilized thereon a plurality of discrete domains each containing a predetermined amount of a reagent capable of binding a component of a binding electrochemiluminescence assay.

35. An apparatus for use in the detection of an analyte by electrochemiluminescence comprising an electrode and a

support having immobilized on a surface thereof a plurality of discrete binding domains each containing a predetermined amount of a reagent capable of binding a component of a binding electrochemiluminescence assay, said electrode and said binding domains being in electrochemical contact.

36. An apparatus as recited in claim 35 wherein the electrode is the working electrode.

37. An apparatus as recited in claim 35 wherein the electrode is the counterelectrode.

38. An apparatus for use in the detection of an analyte by electrochemiluminescence comprising:

- (a) an electrochemical cell
- (b) a first surface containing an electrode
- (c) a second surface having immobilized thereon a plurality of binding domains each containing a binding reagent capable of binding a component of a binding electrochemiluminescence assay, said first and second surfaces being spatially aligned to permit electron transfer between said electrode and an electrochemiluminescence label directly or indirectly linked to a said binding reagent.

39. An apparatus for use in the detection of an analyte by electrochemiluminescence comprising an electrochemical cell and an electrode having immobilized thereon a plurality of binding domains each containing a binding reagent capable of binding a component of a binding electrochemiluminescence assay.

40. A method for conducting a plurality of electrochemiluminescence assays comprising the steps of:

- (a) contacting a plurality of discrete binding domains with a sample containing one or a plurality of analytes of interest under assay conditions;
- (b) causing one or more of said plurality of domains to electrochemiluminescence; and
- (c) detecting electrochemiluminescence at a plurality of said binding domains.

41. A method for conducting a plurality of electrochemiluminescence assays for a plurality of different analytes of interest comprising the steps of:

- (a) contacting a plurality of discrete binding domains, with a sample containing a plurality of analytes of interest under assay conditions;
- (b) causing said plurality of discrete binding domains to simultaneously electrochemiluminescence; and
- (c) simultaneously detecting electrochemiluminescence from each of said discrete binding domains.

42. An article comprising a plurality of discrete binding domains on a support, said binding domains having a relative spatial organization with respect to one another and having different binding specificities for binding a plurality of different analytes of interest in an electrochemiluminescence assay.

43. An article comprising a plurality of discrete binding domains on a support each of said domains having a relative spatial organization with respect to one another and

each having a different binding specificity for an analyte of interest in an electrochemiluminescence assay, each domain containing a different binding component bound to a different analyte of interest.

44. An article comprising a plurality of discrete binding domains on a support, said domains having a relative spatial organization with respect to one another and each having a different binding specificity for an analyte of interest in an electrochemiluminescence assay, each domain being bound directly or indirectly, to a different analyte of interest and to a moiety capable of electrochemiluminescence.

45. An article comprising a plurality of discrete binding domains on a support comprising a porous material each of said domains having a relative spatial organization with respect to one another and each having a different binding specificity for an analyte of interest in an electrochemiluminescence assay each domain containing a different binding component bound to a different analyte of interest.

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46. An article as recited in claim 45, wherein said porous material comprises carbon.

47. An article as recited in claim 45 which said porous material comprises fibrils.

48. An article comprising a plurality of discrete binding domains on a support comprising a functionalized fibril, said domains having a relative spatial organization with respect to one another and each having a different binding specificity for an analyte of interest in an electrochemiluminescence assay,

each domain being bound directly or indirectly, to a different analyte of interest and to a moiety capable of electrochemiluminescence.

49. An article comprising a plurality of binding domains on a support comprising one or more polymer matrices, said domains having a relative spatial organization with respect to one another and each having a different binding specificity for an analyte of interest in an electrochemiluminescence assay, each domain being bound directly or indirectly, to a different analyte of interest and to a moiety capable of electrochemiluminescence.

50. A cassette for use in the detection of an analyte in a sample by electrochemiluminescence comprising:

- (a) a plurality of discrete binding domains on a support; and
- (b) one or more pairs of electrodes and counterelectrodes.

51. A cassette for using the detection of an analyte in a sample by electrochemiluminescence comprising:

- (a) a plurality of discrete binding domains on a support;
- (b) one or more pairs of electrodes and counterelectrodes spatially aligned with said discrete binding domains; and
- (c) means for sample delivery onto said plurality of discrete binding domains.

52. A cassette as recited in claim 49 wherein said plurality of discrete binding domains forms at least one surface capable of binding a component of a binding electrochemiluminescence assay.

53. A cassette as recited in claim 50 wherein said plurality of binding domains includes binding domains having different binding specificities to provide for simultaneous binding of a plurality of different analytes of interest present in a sample.

54. A cassette for detecting or measuring electrochemiluminescence comprising:

- (a) a first support having a plurality of discrete binding domains thereon;
- (b) a plurality of electrode and counterelectrode pairs, each of said plurality of discrete binding domains being aligned with an in proximity to one of said plurality of electrode and counterelectrode pairs, said discrete binding domains and electrode and counterelectrode pairs forming a plurality of cells for detecting or conducting measurements of electrochemiluminescence, said electrode/counterelectrode pairs being addressable by a source of electrical energy in the form of a voltage waveform effective to trigger electrochemiluminescence; and
- (c) means for sample delivery onto said plurality of discrete binding domains.

55. A cassette as recited in claim 53 wherein said plurality of binding domains includes binding domains having

different binding specificities to provide for simultaneous binding of a plurality of different analytes of interest present in a sample.

56. A cassette for detecting or measuring an analyte of interest in a sample comprising:

(a) a first support having a plurality of discrete binding domains on a surface thereof, at least one of said discrete binding domains being of different binding specificity than the other binding domains, each of said plurality of discrete binding domains being hydrophilic and surrounded by hydrophobic regions;

(b) a second support having a plurality of hydrophilic domains comprising reaction media suitable for conducting a chemical assay thereon; and

(c) means for bringing said plurality of discrete binding domains and said plurality of reaction media into contact so that a sample to be analyzed present on each binding domain is contacted with a reaction medium.

57. A cassette as recited in claim 50 in which said discrete binding domains further comprise an internal control.

58. A method for preparing a plurality of discrete binding domains on a support containing binding reagents capable of binding to analytes of interest, comprising the steps of

(a) forming a self assembled monolayer on a support, said monolayer including a linking group A on the monolayer surface not adjacent to the support, said first linking

group A being capable of specifically binding to a linking group B; and

(b) contacting said first linking group A with a binding reagent capable of binding to an analyte of interest, said binding reagent being bound to said linking group B, so that said binding reagent is linked to said monolayer by an A:B linkage to form a binding surface, said binding surface organized as a plurality of discrete binding domains.

59. A process according to claim 57 wherein a plurality of different binding reagents are bound to the plurality of discrete binding domains and wherein said contacting step is carried out by delivering a plurality of fluid samples, each fluid sample including a different binding reagent, onto said monolayer from a plurality of fluidic guides.

60. A method for detecting or measuring an analyte in an electrochemiluminescence binding assay comprising the steps of:

(a) contacting a plurality of discrete binding domains immobilized on a surface of one or more supports with a sample containing a plurality of analytes and a component of said assay linked to an electrochemiluminescence label;

(b) applying a voltage waveform effective to trigger electrochemiluminescence at one or more of said domains in the presence of a reaction medium suitable for conducting an electrochemiluminescence assay; and

(c) detecting or measuring electrochemiluminescence from said plurality of domains.

61. A kit for use in performance, of a plurality of electrochemiluminescence assays for a plurality of analytes of interest comprising:

(a) an article comprising a plurality of discrete binding domains on a support, said binding domains having a relative spatial organization with respect to one another and having different binding specificities for a plurality of different analytes of interest in an electrochemiluminescence assay; and

(b) a vessel containing a reagent necessary for the conduct of said assays.

Sub 62 61. A kit as recited in claim 60 further including an apparatus for performing said assays.

63. A kit for use in performance, of a plurality of electrochemiluminescence assays for a plurality of analytes of interest comprising:

a vessel containing binding components specific for a plurality of different analytes for use in a plurality of electrochemiluminescence assays.

64. A kit as recited in claim 62 further containing one or more vessels containing non-binding components for a plurality of electrochemiluminescence assays.

65. A kit as recited in claim 62 further containing an article comprising a plurality of discrete binding domains on a support, said binding domains having a relative spatial organization with respect to one another and having different

binding specificities for a plurality of different analytes of interest in an electrochemiluminescence assay.

66. An apparatus for measuring electrochemiluminescence of a sample comprising:

(a) a plurality of cells for holding at least one sample, said plurality of cells being formed from a plurality of electrode and counterelectrode pairs and a first support that comprises a plurality of discrete binding domains, said plurality of discrete binding domains being aligned with and in proximity to the plurality of electrode and counterelectrode pairs, said electrode and counterelectrode pairs being separately addressable, said cells being suitable for conducting measurements of electrochemiluminescence,

(b) voltage control means adapted to apply a controlled voltage waveform to said plurality of electrode and counterelectrode pairs, said voltage waveform being effective to trigger electrochemiluminescence in said plurality of cells, and

(c) photon detector means for detecting electrochemiluminescence from said sample.

67. A cassette for detecting or measuring an analyte of interest in a sample comprising

(a) a first support having a plurality of discrete binding domains on the surface thereof to form at least one binding surface, at least some of said discrete binding domains being of different binding specificities than other binding domains, each of said plurality of discrete binding domains being hydrophilic and surrounded by hydrophobic regions and

(b) a second support having a plurality of hydrophilic domains comprising reaction media suitable for conducting a chemical assay thereon to form an assay surface, said plurality of discrete binding domains and said plurality of said reaction media being capable of being brought into contact so that a sample to be analyzed present on each binding domain is contacted with a reaction medium to detect or measure an analyte of interest.

68. A method for detecting or measuring electrochemiluminescence in a sample comprising

(a) contacting one or more of a plurality of discrete binding domains, said plurality of binding domains situated on a surface of one or more supports, with a sample comprising molecules linked to an electrochemiluminescent label, wherein a plurality of electrode and counterelectrode pairs are spatially aligned with said plurality of discrete binding domains, wherein said plurality of electrode and counterelectrode pairs is present on a second support surface,

(b) placing said second support surface proximate to said binding surface so that each said electrode and counterelectrode pair is proximate to a different binding domain,

(c) applying a voltage waveform effective to trigger electrochemiluminescence at each of said plurality of electrode and counterelectrode pairs, and

(d) detecting or measuring said electrochemiluminescence.

69. A method for detecting or measuring analytes of interest in a sample comprising

(a) placing drops of a sample containing an analyte to be detected or measured on a plurality of discrete binding domains on a support surface, said plurality of discrete binding domains comprising at least one binding domain that contains binding reagents that are identical to each other and that differ in specificity from the binding reagents contained within other binding domains, each of said discrete binding domains being characterized as either hydrophobic or hydrophilic, with the proviso that the region of said support surface surrounding each said binding domain is (i) hydrophobic if said binding domain is hydrophilic, and (ii) hydrophilic if said binding domain is hydrophobic, so as to allow said one or more analytes of interest in the sample to bind to said binding domains, and

(b) contacting said drops on said first support with a surface of a second support having a plurality of discrete hydrophilic domains comprising reaction media suitable for conducting a chemical assay thereon, and

(c) determining the presence of said analytes of interest that are bound to said binding domain.

70. A method for detecting or measuring electrochemiluminescence in a sample comprising the following steps in the order stated:

(a) contacting a sample with a surface of a support, said surface containing a plurality of discrete binding

domains, said binding domains being spatially aligned with and capable of being brought into proximity to a plurality of electrode and counterelectrode pairs;

(b) bringing said binding domains into proximity to said plurality of electrode and counterelectrode pairs;

(c) applying a voltage waveform effective to trigger electrochemiluminescence; and

(d) detecting or measuring electrochemiluminescence.

71. A method of detecting or measuring an analyte of interest in a sample comprising:

(a) contacting a sample with a surface of a support, said surface containing a plurality of discrete binding domains, said binding domains being spatially aligned with and capable of being brought into proximity to a plurality of electrode and counterelectrode pairs, said binding domains being capable of binding an analyte of interest, in which said contacting is under conditions such that binding of any analyte in the sample to said binding domains can occur;

(b) contacting said binding domains with a binding partner of the analyte of interest, in which said binding partner is linked to an electrochemiluminescent label;

(c) bringing said binding domains into proximity to said plurality of electrode and counterelectrode pairs;

(d) applying a voltage waveform effective to trigger electrochemiluminescence; and

(e) detecting or measuring electrochemiluminescence, in which an increase in electrochemiluminescence over background levels indicates the presence or amount of the analyte in the sample.

72. A method for conducting a plurality of electrochemiluminescence assays comprising the steps of:

(a) contacting a plurality of discrete polymer matrices, each polymer matrix comprising one or more binding domains with a sample containing one or a plurality of analytes of interest;

(b) causing one or more of said plurality of domains to electrochemiluminesce;

(c) detecting electrochemiluminescence.

73. A method as recited in claim 72 wherein electrochemiluminesce is detected by photographic film.

74. A method for conducting a plurality of electrochemiluminescence assays comprising the steps of:

(a) contacting a plurality of binding domains comprising oligonucleic acids with a sample containing one or a plurality of analytes of interest;

(b) causing one or more of said plurality of domains to electrochemiluminesce; and

(c) detecting electrochemiluminescence at a plurality of said binding domains.

75. A method as recited in claim 74 wherein said plurality of binding domains contain oligonucleic acids linked to electrochemiluminescent labels.

76. A method as recited in claim 74 wherein said plurality of binding domains contain oligonucleic acids linked to electrochemiluminescent labels and one or more of said analytes, when bound to said oligonucleic acids, causes modulation of the electrochemiluminescent signal from one or more binding domains.

77. A method for conducting a plurality of electrochemiluminescence assays comprising detecting a plurality of analytes present in a multicomponent sample at a concentration below about 10^{-3} comprising the steps of:

(a) contacting a plurality of discrete binding domains with a sample containing one or a plurality of analytes of interest;

(b) causing one or more of said plurality of domains to electrochemiluminesce; and

(c) detecting electrochemiluminescence at a plurality of said binding domains.

78. A system for conducting a plurality of electrochemiluminescence assays comprising:

(a) a plurality of binding domains specific for a plurality of different analytes;

(b) a voltage waveform generator; and

(c) a photon detection means.

79. A system for conducting a plurality of electrochemiluminescence assays comprising:

(a) an electrode;

(b) a counterelectrode;

(c) a plurality of binding domains specific for a plurality of different analytes;

(d) means for triggering electrochemiluminescence; and

(e) means for electrochemiluminescence detection.

